Derivation of relaxation induced by spectral diffusion

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(Dated: March 10, 2008)

Abstract

Here is the way to derive Eq. (4.17)

PACS numbers: 71.23.Cq, 72.70.+m, 72.20.Ee, 72.80.Sk

I. TRANSITIONS STIMULATED BY THE SPECTRAL DIFFUSION

This essentially formal section is dedicated to the derivation of Eq. (4.17). This result is quite universal and independent of the particular mechanism of the relaxation. One can briefly explain this result using the following qualitative picture of resonant tunneling.

In this section we calculate the rate of transitions induced by the spectral diffusion assuming that the single two-level system is characterized by the tunneling amplitude Δ_0 and the level shift $\Delta(t)$, which depends on the time and passes through zero due to the spectral diffusion. This condition is satisfied when the characteristic energy shift is less than the dispersion of the interaction $W_T = U_0 P_0 T$. Since fluctuations of the molecular field are caused by the rapid phonon-induced transitions of environmental thermal TLS one can treat the field $\Delta(t)$ classically. One can also assume that the field $\Delta(t)$ is distributed continuously and almost uniformly within the domain $(-W_T, W_T)$ with the distribution density $1/W_T$. The continuity is due to the long-range character of the interaction. It can be shown that any interaction decreasing with the distance slower than exponentially leads to the smooth continuous distribution of possible values of the field $\Delta(t)$ due the thermal fluctuations of thermal TLS. Indeed, the characteristic energy discreteness for the possible values of $\Delta(t)$ induced by the interaction within the radius R scales as $\delta_R \sim \exp(-\eta n_T R^3)$ (where $\eta \sim 1$ is the numerical factor and $n_T P_0 T$ is the density of thermal TLS, while the interaction at longer distances is of order of $1/R^3$ and it will contemporary reduce the discreteness scale filling the gaps in the distribution with increasing the interaction radius. One should note that our consideration is analogous to the work by Prokofev and Stamp [1] where the similar problem has been resolved for the tunneling of electronic spin coupled to the nuclear spin bath and Eq. (11) is quite similar to their final result.

Thus the problem can be reduced to the two levels coupled by the small tunneling amplitude Δ_0 and interacting with the random time-dependent field $\Delta(t)$. We can represent the system Hamiltonian using pseudopin 1/2 model, where the projections $S^z = \pm 1/2$ corresponds to two states of the system and S^x-term is responsible for tunneling

$$\widehat{H} = -\Delta(t) S^z - \Delta_0 S^x. \tag{1}$$

We assume that our system initially occupies the state $S^z = 1/2$ and study the probability of its transfer to the other state $S^z = -1/2$ with the time t. Then the wavefunction can be expressed as

$$c_+(t)|+> + c_-(t)|-> \tag{2}$$

where indexes + and − denotes the states with the projections $S^z = \pm 1/2$, respectively, and the coefficients c_+ and c_- satisfy the time-dependent Schrodinger equation (we set $\hbar = 1$)

$$\begin{aligned} i\frac{dc_+}{dt} &= \frac{\Delta(t)}{2}c_+ + \frac{V}{2}c_-; \\ i\frac{dc_-}{dt} &= -\frac{\Delta(t)}{2}c_- + \frac{V}{2}c_+; \end{aligned} \tag{3}$$

with initial conditions

$$c_+(0) = 1;\ c_-(0) = 0. \tag{4}$$

We are interested in probabilities to find our system in $|+>$ of $|->$ states. These probabilities are expressed by squared absolute values of wavefunction amplitudes $P_+ =| c_+ |^2$, $P_- =| c_- |^2$. Using the Schrodinger equation Eq. (3) one can write down equations for those probabilities as

$$i\frac{d \mid c_+ \mid^2}{dt} = -i\frac{d \mid c_- \mid^2}{dt} = \frac{\Delta_0}{2}(c_-c_+^* - c_+c_-^*). \tag{5}$$

Similarly one can derive the equation for the terms in the right hand side of Eq. (5)

$$\begin{aligned} i\frac{dc_+c_-^*}{dt} &= \Delta(t)c_+c_-^* + \frac{\Delta_0}{2}(1 - 2P_+), \\ c_+^*c_- &= (c_+c_-^*)^*. \end{aligned} \tag{6}$$

Deriving the latter equation we used the conservation of the normalization of our wavefunction $| c_+ |^2 + | c_- |^2 = P_+ + P_- = 1$. One can formally express the left hand side of Eq. (6) in terms of the probability P_+ as

$$\begin{aligned} c_+c_-^*(t) = i\frac{\Delta_0}{2}\int_0^t d\tau \exp\left(i\int_\tau^t d\tau_1\Delta(\tau_1)d\tau_1\right) \\ \times(1 - 2P_+(\tau)). \end{aligned} \tag{7}$$

Substituting this expression into Eq. (5) we get the closed equation for the probability to find our system in the state $S^z = 1/2$

$$\begin{aligned} \frac{dP_+}{dt} &= \\ -\frac{\Delta_0^2}{2}\int_0^t d\tau \cos\left(\int_\tau^t d\tau_1\Delta(\tau_1)d\tau_1\right)(1 - 2P_+(\tau)). \end{aligned} \tag{8}$$

Integrating Eq. (8) with respect to the time we got

$$P_+(t) = 1 - \\ -\frac{\Delta_0^2}{2}\int_0^t dt_1\int_0^{t_1} d\tau \cos\left(\int_\tau^t d\tau_1\Delta(\tau_1)d\tau_1\right)(1-2P_+(\tau)). \tag{9}$$

At this point we are going to study the behavior of $P_+(t)$ using the iteration method. In the first step one can set $P_+^{(0)}(t) = 1$ in accordance with the initial conditions and substitute it to the right hand side of Eq. (9) which leads to the first iteration

$$P_+^{(1)}(t) = 1 \\ -\frac{\Delta_0^2}{2}\int_0^t dt_1\int_0^{t_1} dt_2 \cos\left(\int_{t_2}^t d\tau_1\Delta(\tau_1)d\tau_1\right). \tag{10}$$

To evaluate the expression for P_+ one should average the right hand side of Eq. (9) over possible realizations of the random field $\Delta(t)$. To perform this averaging one can use the fact that the relaxation of the field due to the interaction of thermal TLS with phonons is relatively slow compared to the decay rate of the average cosine $< cos(\Delta t) >$ with the time t due to the static fluctuations of the energy Δ. Indeed, the characteristic dispersion of the random field W_T is proportional to the thermal energy T $W_T \approx U_0P_0T$, while the rate of phonon stimulated transitions in the environment scales as $1/\tau_{ph} \sim AT^3$. Then averaging the cosine function under the integral in Eq. (10) one can approximately assume $\Delta(t) = const$ and perform averaging over various static realizations of $\Delta(t)$ because in this case the average cosine $< cos(\Delta\tau) >$ decays at times $\tau \sim 1/W_T \ll \tau_{ph}$. Then one can replace the integral over τ with the δ-function and we get

$$P_+(t)^{(1)} = 1 - k_1 t; \\ k_1 = \frac{\pi\Delta_0^2 < \delta(\Delta) >}{2} \sim \frac{\Delta_0^2}{W_T}. \tag{11}$$

This equation leads to the estimate of the transition time reproducing the earlier used expression Eq. (11). Its accuracy can be verified considering the next iteration in Eq. (9).

$$P_+^{(2)}(t) = 1 \\ -\frac{\Delta_0^2}{2}\int_0^t dt_1\int_0^{t_1} dt_2 \cos\left(\int_{t_2}^{t_1} d\tau_1\Delta(\tau_1)d\tau_1\right) \\ +\frac{\Delta_0^4}{4}\int_0^t dt_1\int_0^{t_1} dt_2 \\ \int_0^{t_2} dt_3\int_0^{t_3} dt_4\left\langle\cos\left(\int_{t_2}^{t_1} d\tau_1\Delta(\tau_1)\right)\cos\left(\int_{t_4}^{t_3} d\tau_2\Delta(\tau_2)\right)\right\rangle \tag{12}$$

Correction terms should be averaged over random realizations of the field $\Delta(t)$. Averaging of the first term yields the answer Eq. (11). The second term is much more complicated. It describes the correction due to the coherent return of the system back to its initial state. If the field Δ is time-independent then this term is very large because of the phase compensation of two cosines. Taking Δ to be constant and performing averaging over its random realization one can get for the main term

$$\delta_{stat}^{(2)} = = \frac{\Delta_0^4}{8}\int_0^t dt_1 \int_0^{t_1} dt_2 \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 F(W_T(t_1 - t_2 - t_3 + t_4)), \tag{13}$$

where

$$F(W_T t) =< exp(i\Delta t) >, \tag{14}$$

and averaging is made over all static realizations of the random field Δ having the characteristic value W_T. Integration over one of the four time arguments in Eq. (13) yields the factor $1/W_T$ similarly to Eq. (11) while integration of remaining constant over all other three arguments results in the factor t^3 so we get

$$\delta_{stat}^{(2)} \approx \frac{\Delta_0^4 t^3}{W_T}. \tag{15}$$

This static field correction becomes comparable with the first order term Eq. (11) at $t \sim 1/\Delta_0$ and after this time we cannot use the first approach. The average transition probability is then given by $P_{-} \sim \mid \Delta_0 \mid /W_T$. This transition probability describes the effect of overlap between two different levels induced by their coupling Δ_0. The transferred density Δ_0/W_T is associated with rare resonances having $|\Delta| \leq |\Delta_0|$ where almost the whole density is transferred. The factor Δ_0/W_T expresses the probability of such resonance which defines the average population transfer.

The situation is quite different when the fields $\Delta(t)$ changes with the time and the memory about its previous value is lost during the characteristic time $\tau_* \approx T_2$ induced by the irreversible interaction with phonons. The correlation time τ_* can be approximated by the system dephasing time T_2 which is evident from the one to one correspondence of the second correction to the probability expression with the standard expression for the spin echo-amplitude caused by the similar phase correlations. For a crude estimate of the correction we make a straightforward assumption that when $t_1 - t_4 > \tau_*$ in Eq. (10) the correlation between two cosines can be totally neglected, while at $t_1 - t_4 < \tau_*$ one can neglect the time-dependence of the field Δ. Then we have two contributions expressed by correlated and non-correlated terms. The correlated term has the form similar to the static field expression Eq. (15) with the important replacement of t^3 factor with the smaller factor $t\tau_*^2$

$$\delta_{corr}^{(2)} \approx \frac{\Delta_0^4}{4}\int_0^t dt_1 \int_{t_1-\tau_*}^{t_1} dt_4 \int_{t_4}^{t_1} dt_2 \cos\left(\int_{t_2}^{t_1} d\tau_1 \Delta(\tau_1)\right) \int_{t_4}^{t_2} dt_3 \cos\left(\int_{t_4}^{t_3} d\tau_2 \Delta(\tau_2)\right) \approx \frac{\Delta_0^4 \tau_*^2 t}{W_T}, \tag{16}$$

while the second term can be well approximated by the second expansion term of the standard exponent describing relaxation

$$\delta_{noncorr}^{(2)} \approx \frac{\Delta_0^4}{4}\int_0^t dt_1 \int_0^{t_1} dt_2 \left\langle \cos\left(\int_{t_2}^{t_1} d\tau_1 \Delta(\tau_1)\right)\right\rangle \int_0^{t_2} dt_3 \int_0^{t_3} dt_4 \left\langle \cos\left(\int_{t_4}^{t_3} d\tau_2 \Delta(\tau_2)\right)\right\rangle \approx \frac{(k_1 t)^2}{8}. \tag{17}$$

It can be shown making independent averaging of cosine in Eq. (9) that the collection of all noncorrelated terms leads to the exponential behavior of the probability function

$$P_+(t) = 1/2 + 1/2(1 - \exp(-k_1 t). \tag{18}$$

The correction to this behavior can be estimated comparing Eq. (16) with the first order correction $\delta_1 \approx k_1 t$ in Eq. (11). This yields

$$\frac{\delta_{corr}^{(2)}}{\delta_1} \approx \Delta_0^2 \tau_*^2. \tag{19}$$

Since for nonadiabatic TLS we took the amplitude Δ_0 small Eq. (4.16) is small compared to $1/T_2$ one would expect that this and other corrections are always very small so one can use Eq. (11) without any restrictions. One should note that the more accurate averaging using the telegraph process formalism as in Refs. [2], where the similar consideration has been applied to the spin system, leads to the same result.

It is useful to discuss the transitions described above qualitatively. Real transitions take place when the field $\Delta(t)$ is small. Assume that there exists the resonant domain $(-\epsilon_*, \epsilon_*)$ and transitions take place mostly when $\Delta(t)$ belongs to this domain. The size of this domain can be defined by the following self-consistent analysis. The size of the domain ϵ_* exceeds extremely small tunneling Δ_0 because the energy change during the time $1/\Delta_0$ is much larger than Δ_0. If the energy change with the time is negligible then the population transfer takes place during the time $t_* \sim \hbar/\epsilon_*$ and after that the coherent oscillations of population take place. The transferred population is given by the standard quantum mechanical expression $P_* \approx \Delta_0^2/\epsilon_*^2$. If the change of energy during the time τ_* exceeds the energy ϵ_* then we cannot use ϵ_* in the denominator of the expression for P_* but the larger energy associated with the energy fluctuation should be used there. One can introduce the minimum energy scale ϵ_* such as the energy fluctuation during the time $\hbar/\epsilon_*$ coincides with this energy. The definition of our energy scale can be made in terms of the decoherence time τ_2 as

$$\epsilon_* = \hbar/T_2, \tag{20}$$

where the decoherence time is the time corresponding to the phase fluctuation $\delta\phi(t) \sim \int_0^t d\tau(\Delta(\tau) - \Delta(0))$ becoming of order of unity. One can estimate this decoherence time setting the product of the energy fluctuation during the time t which is $\delta(t) \sim W_T(t/\tau_{ph})$ (see Refs. [2]) and the time t to be equal unity. This yields

$$\epsilon_* = \frac{\hbar}{\tau_2} \sim \left(\frac{W_T\hbar}{\tau_{ph}}\right)^{1/2}. \tag{21}$$

When the transition energy Δ exceeds ϵ_* the transferred population during the time of energy change less or of order of Δ behaves as Δ_0^2/Δ^2 so we can neglect these contributions compared to the resonant contribution. Thus transitions take place mostly when energy Δ enters the resonant domain and for each entrance the transition probability is given by $(\Delta_0/\epsilon_*)^2$. During the time $t \gg \tau_{ph}$ the system can enter to the resonant domain $P_t \approx t/\tau_2$ times and the probability to enter there P_ϵ is given by the ratio of the resonant domain size ϵ_* to the whole energy domain size $P_\epsilon \approx \epsilon_*/W_T$. The total probability of transition during the time t is given by the product of all three factors $P_* \cdot P_t \cdot P_\epsilon$ leading to the phonon independent result $k_1 t \sim \Delta_0^2 t/W_T$. Setting this probability to unity one can estimate the transition rate as $k_1 \sim \Delta_0^2/W_T$ in agreement with Eq. (11).

[1] N. V. Prokofev, P. S. Stamp, review.

[2] A. L. Burin, L. A. Maksimov, K. N. Kontor, Theor. Math. Physics (1992).